Faraday rotation angle anisotropy in a Fe-based diluted magnetic semiconductor

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The magnetic-field and temperature dependences of the Faraday rotation angle in the cubic crystal $Cd_{1-x}Fe_xTe$ has been investigated theoretically and experimentally. It is shown that by simultaneously taking into account the spin orbit and Zeeman interactions without use of perturbation theory the anisotropic nature of these dependences in strong magnetic fields can be explained in a unified approach. © *1997 American Institute of Physics.*
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Semimagnetic semiconductors (SMSCs) containing iron, a magnetic component, are characterized by a number of distinguishing features. One is the manifestation of an anisotropic character of the magnetization, the anisotropy becoming especially large in strong magnetic fields at liquid-helium temperature. As shown in Ref. 1, this in turn results in an anisotropy of the Faraday effect in the cubic crystal $Cd_{1-x}Fe_xTe$. There exist different approaches for explaining the magnetization anisotropy in this material in strong magnetic fields.[2,3]

In the paper we report the results of an experimental and theoretical study of the Faraday effect in $Cd_{1-x}Fe_xTe$. Measurements of the Faraday rotation angle were performed on samples with Fe concentration $x=0.03$, which were preoriented along the [100] and [111] crystallographic axes, at temperatures in the range 4.2–100 K. A ~20% anisotropy of the temperature dependence of the Verdet constant was observed in strong magnetic fields up to 200 kG.

$Cd_{1-x}Fe_xTe$ ($x\leqslant 0.05$) single crystals were grown by a modified Bridgman method and the method of horizontally directed crystallization. The composition of the solid solutions was fixed by the charge of the initial components and checked by atomic-absorption and microprobe analysis. The composition of the samples employed was additionally checked by observing excitonic structure in the reflection spectra of the crystals at 4.2 K. Samples employed in the magneto-optic investigations consisted of (100) and (111) wafers with thickness $d=0.2-3.0$ mm.

The Faraday rotation angle (θ_F) measurements were performed in strong magnetic fields with maximum intensity up to 200 kOe, produced by a pulsed magnet consisting of a copper solenoid with an inside diameter of 6 mm and a capacitor bank with total capacitance $C=2400$ μF and working voltage up to 5 kV. The experimental sample was mounted at the center of the solenoid together with a magnetic test coil and a Ge sensor for monitoring the field intensity H and the temperature, respectively. A helium optical cryostat and a UTREKS temperature regulation system were used to conduct measurements in the temperature range 5–295 K.

The results of the measurements of the magnetic-field dependence of the Faraday rotation angle for different directions of the magnetic field are presented in Fig. 1. The temperature dependence of the Verdet constant is shown in Fig. 2. It was determined that the anisotropy of the angle θ_F and the Verdet constant V at low temperatures and in fields ~200 kG is approximately 20%.

To investigate this problem theoretically, we proceed from the fact that the ground state of an isolated Fe^{2+} ion, which possesses a $3d^6$ configuration, is the term ^5D. In a crystal field with T_d symmetry the term splits into a ^5E orbital doublet and a $^5T_2$ orbital triplet. The Hamiltonian of the Fe^{2+} ion in an external magnetic field, taking into account the spin-orbit interaction, is

$$H=H_0+H_{CF}+\lambda\mathbf{L}\cdot\mathbf{S}+\mu_B\mathbf{B}\cdot(\mathbf{L}+2\mathbf{S}), \qquad (1)$$

where H_0 is the Hamiltonian of the isolated atom, H_{CF} takes

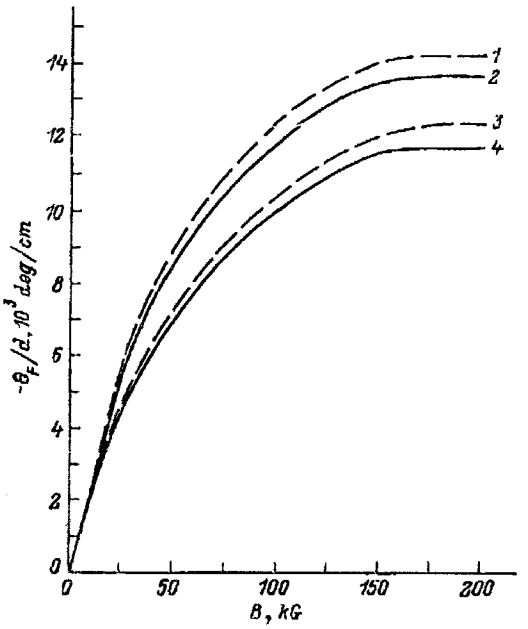

FIG. 1. Faraday rotation angle versus magnetic field in $Cd_{1-x}Fe_xTe$ ($x=0.03$) at temperature $T=5$ K with photon energy $E=1.459$ eV and different orientations of **B** relative to the crystallographic axes. *2*, *4* — experimental results for [100] and [111] orientations, respectively; *1*, *3* — computational results for the same orientations.

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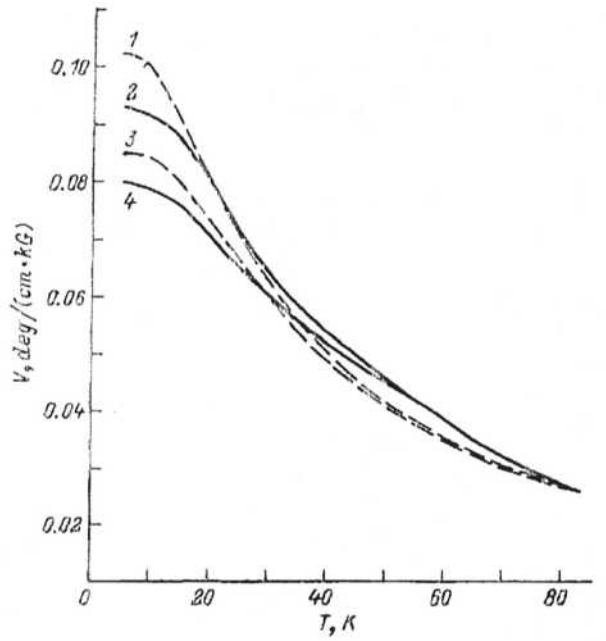

FIG. 2. Temperature dependence of the Verdet constant V in $Cd_{1-x}Fe_xTe$ ($x=0.03$) in a magnetic field $B=135$ kG and photon energy $E=1.459$ eV for different orientations of B with respect to the crystallographic axes. *2*, *4* — Experimental results for [100] and [111] orientations, respectively; *1*, *3* — computational results for the same orientations.

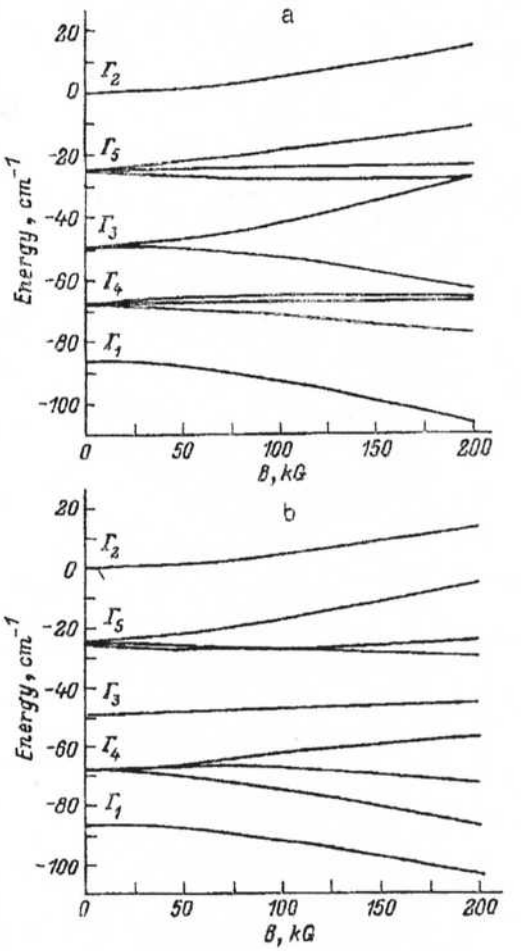

FIG. 3. Magnetic field dependence of the spin-orbit levels which originates from the orbital term E for $\mathbf{B} \parallel [100]$ (a) and $\mathbf{B} \parallel [111]$ (b).

into account the crystal field, and λ is the spin-orbit interaction constant. In the approximation[4] of equivalent operators, H_{CF} is given by

$$H_{CF}=A\left[\frac{1}{8}(L_+^2+L_-^2)^2+\frac{3}{2}L_z^4-6L_z^2-\frac{12}{5}\right],$$

$$L_\pm=L_x\pm iL_y, \tag{2}$$

where A is a constant characterizing the interaction of the impurity ion Fe with its crystalline environment. The basis functions of the orbital terms ^5E and $^5T_2$ have the form[4]

$$\begin{aligned}
&^5E: \quad u_1=|0\rangle,\\
&\qquad u_2=(1/\sqrt{2})(|2\rangle+|-2\rangle);\\
&^5T_2: \quad v_1=|-1\rangle,\\
&\qquad v_2=(1/\sqrt{2})(|2\rangle-|-2\rangle),\\
&\qquad v_3=-|1\rangle.
\end{aligned} \tag{3}$$

In the absence of an external magnetic field the spin-orbit interaction splits the orbital doublet ^5E in second-order perturbation theory into the levels Γ_1, Γ_4, Γ_3, Γ_5, and Γ_2; the energy splitting between them $6\lambda^2/\Delta$ is ~ 24 cm^{-1} (for Fe $\Delta=6A=2500$ cm^{-1} and $\lambda=-100$ cm^{-1}). In strong magnetic fields $B\simeq 150$ kG, $\mu_B B\simeq 10$ cm^{-1}. Since the energy splittings presented are of the same order of magnitude, the spin-orbit and Zeeman interactions must be taken into account simultaneously.

In the basis (3) the Hamiltonian (1) has the form

$$H=\begin{bmatrix} E_0(^5E)+2\mu_B B\mathbf{n}\cdot\mathbf{S} & (\lambda\mathbf{S}+\mu_B B\mathbf{n})\cdot\mathbf{U} \\ (\lambda\mathbf{S}+\mu_B B\mathbf{n})\cdot\mathbf{U}^+ & E_0(^5T_2)+2\mu_B B\mathbf{n}\cdot\mathbf{S}-(\lambda\mathbf{S}+\mu_B B\mathbf{n})\cdot\mathbf{I} \end{bmatrix}, \tag{4}$$

where $E_0(^5E)$ and $E_0(^5T_2)$ are the eigenvalues of H_{CF} ($E_0(^5T_2)-E_0(^5E)=\Delta$), $\mathbf{n}$ is a unit vector in the direction of the magnetic field, and

$$\mathbf{U}_{\nu k}=\langle u_\nu \mathbf{L} \nu_k\rangle, \quad \mathbf{I}_{kk'}=-\langle \nu_k \mathbf{L} \nu_{k'}\rangle. \tag{5}$$

Substituting the explicit form of the matrix elements (5) with respect to the basis (3) and using the spin functions

($M_s = -2, -1, 0, 1, 2$), we obtain a 25×25 matrix of the Hamiltonian (4). This approach does not employ perturbation theory and makes it possible to take into account the mixing of the ^5E and $^5T_2$ terms. The magnetic-field dependence of the spin-orbit terms, which were obtained from the orbital doublet ^5E, is shown in Fig. 3.

We shall calculate the Faraday rotation angle according to Ref. 5:

$$\theta_F = \frac{F_0^{1/2} d}{2\hbar c} \frac{E^2}{(E_0^2 - E^2)^{3/2}} \frac{(J_h - J_e)}{g\mu_B} M. \quad (6)$$

Here J_e and J_h are the exchange interaction integrals for electrons and holes with the angular momenta of the Fe ions; E_0 is the excitonic transition energy; E is the photon energy; F_0 is a constant, which contains the oscillator strength of the excitonic transition; g is the g-factor of the Fe^{2+} ion; M is the magnetization per unit volume

$$M = x k_B T \frac{\partial}{\partial B} \ln Z, \quad (7)$$

x is the concentration of Fe^{2+} ions; and Z is the partition function, which is found with the aid of the energy spectra obtained in a magnetic field (Fig. 3).

In Figs. 1 and 2 the computational results for the magnetic-field and temperature dependences of the Faraday rotation angle are compared with the experimental data. As one can see from these curves, the experimental and theoretical results are in satisfactory agreement with one another. The Faraday rotation anisotropy is due to the characteristic behavior of the spin-orbit levels in strong magnetic fields (Fig. 3). The lower level Γ_1 is weakly anistropic, the magnitude of the splitting of the next level Γ_4 is substantially different for magnetic field orientations $\mathbf{B} \parallel [100]$ and $\mathbf{B} \parallel [111]$, and the Γ_3 level remains doubly degenerate with field orientation $\mathbf{B} \parallel [111]$, while in a field $\mathbf{B} \parallel [100]$ it is appreciably split.

We note that different variants of the perturbation theory, with allowance of the mixing of the different orbital levels and disregarding them in order to explain the anisotropy of the magnetization, were used in Refs. 4, 6, and 7. However, the energy spectrum obtained, for example, in Ref. 4, cannot be used to explain the indicated features from a unified standpoint. Taking into account simultaneously the spin-orbit and Zeeman interactions is therefore justifiable, this makes it possible to give a unified description of the Faraday rotation angle anisotropy and the characteristic features of the low-temperature dependence of the Verdet constant.

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